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PICATINNY ARSENAL TECHNICAL REPORT 3157

INADEQUACY OF THERMAL INITIATION
THEORY IN INTERPRETING RESULTS OF
FISSION - FRAGMENT IRRADIATION OF
EXPLOSIVES AT ELEVATED TEMPERATURES

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JUNE 1964

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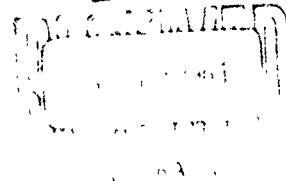
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**INADEQUACY OF THERMAL INITIATION THEORY
IN INTERPRETING RESULTS OF FISSION-FRAGMENT
IRRADIATION OF EXPLOSIVES AT ELEVATED TEMPERATURES**

by

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June 1964

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Technical Report 3157

AMCMS Code 5011 11.818A

DA Proj 1C010501A007

Approved

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ABSTRACT

An extreme test of the "hot-spot" model of explosive initiation as applied to microscale irradiation events should occur when the explosives are irradiated at elevated temperatures. To so test this model, fission-fragment irradiation of four explosives selected for their high reaction rates (PDX, HMX, PETN, and nitroglycerin) were performed at 125° to 215 °C. No explosions or signs of thermal decomposition were observed with any of the explosives. Since a detailed analysis of the resulting cylindrical zones of radiation heating and their subsequent behavior with time predicts initiation for these particular explosives, it appears that the "hot-spot" model is inadequate in describing microscale irradiation phenomena.

INTRODUCTION

Considerable research has been done in recent years (Ref 1) to determine the basic conditions necessary for the initiation of an explosive. Most of the more extensive studies have been made on the macroscopic behavior of explosives and have been interpreted satisfactorily in terms of the classical "hot-spot" model (Refs 2-6). Several explosives have also been subjected to irradiation, but not until recently has a detailed analysis of this process been attempted.

Cerny and Kaufman (Ref 7) irradiated several explosives with pions to produce spherical hot spots less than 10^{-4} cm in diameter. In this work it was noted that no initiations had occurred. However, it was shown that the high energy-density produced by pion capture (when assumed to quickly degrade to thermal energy) (Refs 8 and 9) should cause a temperature increase sufficient to initiate several of the irradiated explosives.

A comparative analysis of typical pion-capture and nuclear-fission events shows that, in the latter process, a higher energy-density occurs and hence the temperature increase is greater. As is discussed in detail elsewhere (Ref 7), the energy deposited by the Auger electrons and alpha particles which arise from a "typical" pion capture event produces a spherical hot spot. The intense ionization and excitation along a fission-fragment track, however, produce a high energy-density region of cylindrical geometry.

Irradiations of a few explosives have been performed with fission-fragments (Refs 10 and 11) but no initiations have been observed. (The explosions of nitrogen iodide are apparently due to a surface effect rather than a thermal spike (Refs 12-14).) However, the calculations of Cerny and Kaufman (Ref 7) indicated that some of the more sensitive secondary explosives had not been investigated and that, surprisingly, the frequently investigated lead azide was not expected to initiate on this model.

The purpose of the present work was to determine the behavior of several of these more sensitive high explosives when irradiated with fission-fragments at a controlled elevated temperature. Table 1 (p 11) shows the temperature required to produce various degrees of decomposition in 10^{-11} seconds (the characteristic time for heat dissipation) for all the explosives investigated. It is apparent that the strength of the thermal spike arising from the irradiation need not be great to produce appreciable heat from chemical decomposition. Then, by having the explosives at some elevated temperature,

say 150°C, the likelihood of an irradiation-induced initiation—according to the "hot-spot" model—should be significantly increased. Table 1 also shows the temperatures used in the experiment. Four high purity (>99%) secondary explosives were investigated: pentaerythritol tetranitrate (PETN), hexahydro-1,3,5-trinitro-s-triazine (RDX), octahydro-1,3,5,7-tetranitro-s-tetrazine (HMX), and glyceryl trinitrate (nitroglycerin).

EXPERIMENTAL PROCEDURE

A variable-temperature explosive container was used to heat the explosive samples. A small quantity (0.30 ml) of an acidic solution containing Cf-252 (3% spontaneous fission) was added to 300 mg of each explosive. In the case of the three solid explosives the mixture for each pellet was then dried, mixed, and pressed onto a 10-mil thermocouple in right-cylindrical pellets approximately one-quarter inch in diameter and one-quarter inch high. Each pellet was inserted in the container and heated on all surfaces except the top from room temperature to approximately 35°C below the melting point of the explosive. The pellet was kept at this temperature for thirty minutes and then heated to ignition. The time to ignition, which ranged from 4 to 10 minutes, and the temperatures of the pellet and the container were recorded. Five pellets of each explosive were studied in this way. The behavior of the explosive pellets containing Cf-252 was compared to that of pellets of the same explosive prepared in an identical manner but without Californium.

The amount of Cf-252 in the pellets was determined by measuring the alpha-activity of small samples of the solution in a liquid scintillation counter. The ratio of spontaneous fission to alpha-decay being known, it was possible to ascertain that at least three fissions per minute occurred in the explosive samples. Table 2 (p 11) shows the parameters of typical light and heavy fission-fragments for Cf-252 (Ref 15).

The liquid nitroglycerin samples were easier to irradiate since it was only necessary to add the solution containing the Cf-252. The mixture was then placed into the container in a small beaker and the thermocouple immersed.

A second method of irradiation also employed was using Cf-252 plated on small platinum foils. In this case, a foil was placed in direct contact with the top of the pellet (immersed in the case of nitroglycerin) and the pellet

was heated as before. Procedures similar to those used in the first method to control heating to ignition and to measure the activity were followed. The amount of radiation to which each pellet was exposed was determined to be 33 fragments per minute, which was appreciably greater than that of the first method.

RESULTS AND DISCUSSION

No differences between the response of the irradiated explosives and the standards were observed even though the former were exposed to about 200 to 2000 fission-fragments. The "hot" pellets showed no signs of accelerated decomposition (compared to the standards) either when held at the elevated temperature or when heated to ignition. Both types of pellets of an explosive ignited at the same temperature, in the same general manner, and in the same time within the normal range of deviation for such measurements of explosives.

Although the fission-fragment irradiation did not initiate these explosives, a detailed quantitative analysis was made to determine the predicted behavior within the framework of the "hot-spot" model (Refs 2-6). This model has enjoyed some success in explaining the behavior of explosives on a macroscopic scale, but has not often (Ref 7) been used in detail in an analysis of microscale processes.

Initial "temperature"-radius profiles for the "hot-spots," which in reality are energy-radius profiles, were obtained by calculating the energy deposited in successive cylindrical shells coaxial with the fission-fragment track. Such calculations are complicated, however, by a lack of precise information on the relative amounts of energy that a stopping fission-fragment loses to excitations and ionizations. Magee (Ref 16) discusses the ionization and excitation resulting from the passage of high-energy charged particles through matter and postulates that two processes—termed "head-on" and "glancing" collisions—make equal contributions to the differential energy loss of the particle. Employing this model for the initial part of its track, we assume that a fission-fragment would lose half of its energy in producing delta rays (knock-on electrons of energy, say, greater than 200 ev) and the other half in low-energy ionizations and excitations. This latter type of energy deposition was assumed to occur within 10A of the center of the track (Ref 17). Although the values chosen for the minimum delta ray energy and the radial distance for lesser energy drops are somewhat arbitrary, it

is evident (Refs 17 and 18) that 200-ev is of the correct order of magnitude for such a figure and that for the explosives investigated 10A is approximately the straight-line distance traveled by a 200-ev electron.

The relative number of delta rays of a given energy that are produced by the fission fragment can be found using the relation of Mott as cited by Bradt and Peters (Ref 19). This expression was normalized (over all possible energies) to one-half the value of (dE/dx) which was calculated from the range-energy curves of Fulmer (Ref 20). This particular (dE/dx) was chosen at the point along the track where the fragment had lost one-half its initial energy.

The electron range-energy relation used to find the energy deposited away from the track was that of Glocker as cited by Katz and Penfold (Ref 21). To account for the fact that such slow electrons do not move out in a straight line nor in a plane perpendicular to the fragment track, one-half of the practical range as given by Glocker was used as an average, or effective, range (Refs 22-25).

The time required for the passage of a fission-fragment and its subsequent energy deposition is of interest. For a section of the track where $l \sim 100A$, the passage of the fragment through this region and the stopping of the low-energy delta rays should occur in appreciably less than 10^{-13} seconds. Hence we may assume that the initial energy distribution arises in 10^{-13} seconds.

For a first-order reaction, the subsequent time-behavior of the (cylindrical) hot spot is governed by the heat diffusion equation:

$$\frac{\partial T}{\partial t} = \kappa \nabla^2 T + \frac{Q}{c} \eta Z e^{-E/RT} \quad (1)$$

where T is the absolute temperature, t is the time, κ is the thermal diffusivity, Q is the heat of reaction, c is the specific heat, η is the mass fraction of unconsumed reactant, Z is the Arrhenius pre-exponential factor, E is the activation energy, and R is the gas constant. Also $\kappa = \frac{k}{\rho c}$, where k is the thermal conductivity and ρ is the density. These constants are listed in Table 3 (p 12) for the explosives investigated.

Solutions to Equation 1 for later times were obtained consistent with the initial temperature profile and subsequent reactant consumption. The method employed was an extension of the Schmidt method, modified to include the heat liberated by chemical reaction (Ref 37). The material was divided into a series of concentric cylindrical shells, the temperature of each shell being uniform, and the temperatures of the various shells determined by the initial temperature-radius profiles (Figs 1-3, pp 13-15). The temperature for the i th shell, T_i , at time $t + \Delta t$ was then obtained from the temperatures at time t by the approximation:

$$T_i(t + \Delta t) = \left[\frac{\kappa}{(\Delta r)^2} \left(1 + \frac{\Delta r}{2 r_i} \right) \left[T_{i-1}(t) - T_i(t) \right] - \frac{\kappa}{(\Delta r)^2} \left(1 + \frac{\Delta r}{2 r_i} \right) \times \left[T_i(t) - T_{i+1}(t) \right] + \frac{Q}{c} n_i(t) Z \right] \Delta t + T_i(t) \quad (2)$$

where Δr is the thickness of each cylindrical shell and r_i is the radius to the center of the i th shell.

We have assumed that, since molecular vibration frequencies are $\sim 10^{12}$ /sec, it is possible to have the initial energy profile transformed into a temperature profile by 10^{-12} seconds. Secondly, we have assumed that this transition from an energy-radius to a temperature-radius distribution can be estimated by using Equation 1, neglecting chemical reaction. After 10^{-12} seconds, the complete solution to Equation 2 is obtained. The temperature-radius profiles were calculated using an IBM 709 digital computer. Equation 2 was solved for a series of time intervals, Δt , being chosen in such a way that the temperature change in any shell during the time interval was less than 5%; the shell thickness, Δr , was taken to be 5Å. The results for RDX, nitroglycerin, and PETN are shown in Figures 1, 2, and 3, respectively, with initiation predicted in all cases. The behavior of HMX is expected to follow very closely that of RDX.

An alternative approach was used to check the sensitivity of the results to a variation in the initial energy-radius profile. In this case, energy deposition was assumed to be due to delta rays alone so that the temperatures for the first three shells (15Å) were essentially identical ($\sim 1.4 \times 10^4$ °C in RDX). These calculations also predicted that initiation should occur in each explosive in much the same manner as in the initial case. Although

initiation is predicted for all the explosives investigated, such is not the case for explosives in general, as was shown previously (Ref 7).

The thermochemical and physical values used in the calculations are listed in Table 3 (p 12). It was assumed that the thermal conductivity and specific heat were temperature independent, and further that the specific heat of the products of reaction was the same as that of the original explosive. Macroscopic thermal parameters were used throughout as in previous work, since no evidence against their suitability had been found (Ref 38).

These results show once more the inadequacy of the "hot-spot" model in describing the effects of microscale events in explosive materials. It should be noted that Bowden and Yoffe (Ref 1) have postulated that a critical size of 10^{-5} to 10^{-4} cm must be exceeded before the model is applicable; however, it is not apparent from the mathematical model itself just how this limitation comes about. Two theories have been proposed recently which when applied to our experimental conditions appear to yield results consistent with ours. A recent estimate has been made by Dodd (Ref 39) using a statistical approach which indicates that simultaneous decomposition of on the order of only 10 molecules is sufficient for initiation to occur. Secondly, a statistical model has also been used by Ling (Ref 40) to describe initiation phenomena. In applying this model to the case of fission-fragment irradiations, he finds that for temperatures of 200-400°C in the volume around the track initiation is unlikely. It appears, however, that if his calculations were extended to include the higher temperatures indicated in this work (say 1500°C in a 100Å radius cylindrical volume), this model would also predict that initiation is likely.

On the other hand, some of the assumptions made in our work are open to question. The extrapolation of kinetic data to the conditions of this experiment is unavoidable but possibly erroneous. Certainly, the processes involved in the conversion of energy to temperature and their interrelation with chemical reaction are not well understood when considered on this time scale (Ref 41). It is conceivable, as has been noted in shock-wave experiments (Refs 42 and 43), that a delay may exist between the time at which a temperature distribution is established and the beginning of chemical reaction. It should also be noted that calculations dealing with atomic displacements in copper indicate that heat dissipation may occur more rapidly in microscale events than would be expected from classical heat conduction (Ref 44). If there is an analogy, such an effect in explosives would considerably alter our calculations. Lacking more precise information

concerning the assumptions made, however, it appears that the "hot-spot" model may require considerable revision to account for experimental data on explosive irradiations.

We wish to thank Dr. Harold J. Matsuguma for his assistance in the preparation of the experiment.

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TABLE 1

Temperatures required for various fractional decompositions*

Explosive	Maximum Possible Temperature Increase (Q/c), °C	Temperature of the Explosive (°C) Required to Produce a Decomposition of			Experimental Temperature, °C
		1%	50%	99%	
RDX	4850	750	935	1035	160
HMX	4020	805	1025	1160	215
PETN	4930	1065	1625	2050	125
Nitroglycerin	5380	640	815	920	180

*These are the temperatures at which each of the explosives must be kept for 10^{-11} seconds in order to produce 1%, 50%, and 99% chemical decomposition. Also listed are the temperature increases that would result from complete decomposition (Q/c), and the temperatures at which the irradiations were performed.

TABLE 2

Typical parameters for fission-fragments
from the spontaneous fission of Cf-252

Parameter	Units	Light	Heavy
Mass (M)	amu	111	141
Energy (E)	Mev	100.5	78.0
Velocity (V)	cm/sec	1.32×10^8	1.03×10^8
Range (R)	μ of RDX	8.4	7.0
Charge (Z)	-----	44	54

TABLE 3
Physical constants of the explosives

Quantity	Units	RDX		Nitroglycerin		PETN		HMX	
		Value	Ref	Value	Ref	Value	Ref	Value	Ref
ρ	g/cm ³	1.66	26	1.596	28	1.46	1	1.66	26**
c	cal/g °C	0.264	26	0.300	29	0.272	29	0.315	34
k	cal/sec cm ² C	4.9×10^{-4}	26	5.03×10^{-4}	29	6×10^{-4}	1	4.9×10^{-4}	26**
Q	cal/g	1280	27	1615	30	1341	27	1266	35***
Z	sec ⁻¹	$10^{11.2}$	26	$10^{11.5}$	31, 32	$10^{11.1}$	33*	$10^{10.7}$	36
E	kcal/mole	57.2	26	48.0	31, 32	38.6	33*	52.7	36
Formula	--	$C_3H_5N_5O_6$	--	$C_3H_5N_3O_9$	--	$C_5H_8N_4O_{12}$	--	$C_4H_8N_8O_8$	--

*Cook and Abegg, recognizing the autocatalytic nature of PETN, extrapolated to find the "unimolecular" (first order) initial rate for the reaction. This of course becomes considerably faster in the presence of its decomposition products.

**The values for the density (ρ) and the thermal conductivity (k) of RDX and HMX were set equal because of their similar chemical character and because consistent data for the pure materials is not available.

***Johnson (Ref 35) cites Q_L for HMX as 1356 cal/g, from "Explosion Effects Data Sheets," NAVORD Report 2896 (NOL), 14 June 1955. (Addendum of 1956). However, Tonegutti (Ref 27) points out that the difference between Q_L and Q_V for RDX is 90 cal/g. Assuming the same relationship for HMX (since Q_L is nearly the same, as are most of the properties), we obtain 1266 cal/g for Q_V for HMX.

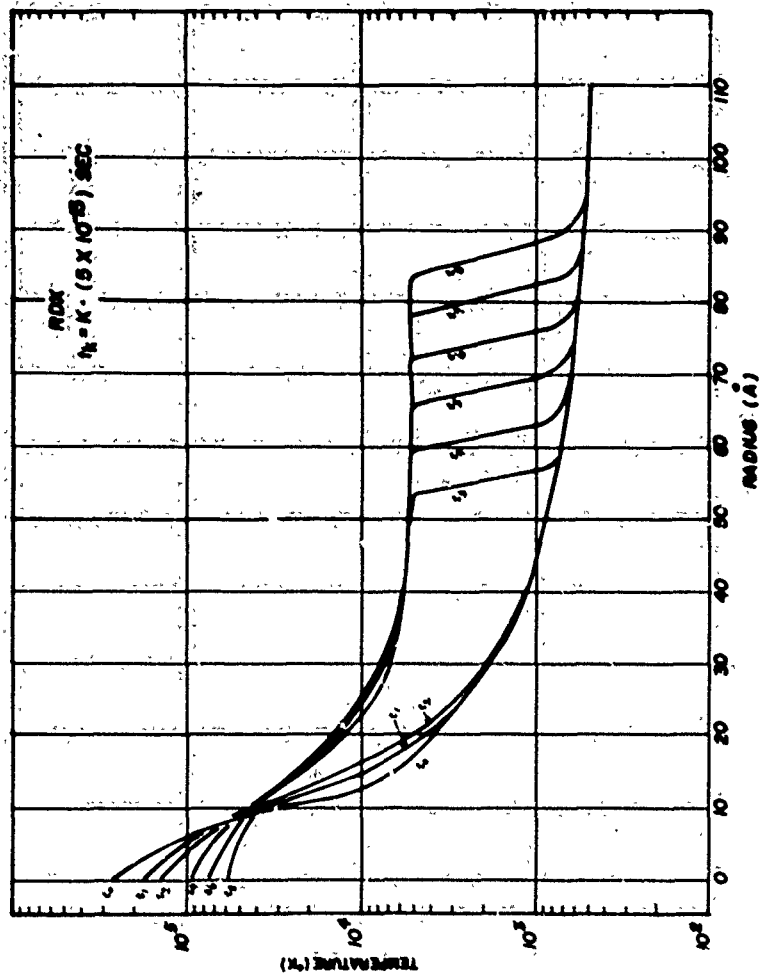


Fig 1. Temperature vs radius profiles for various times following the passage of the fission-fragment through RDX. t_k represents a time period of k times the interval 5×10^{-11} seconds

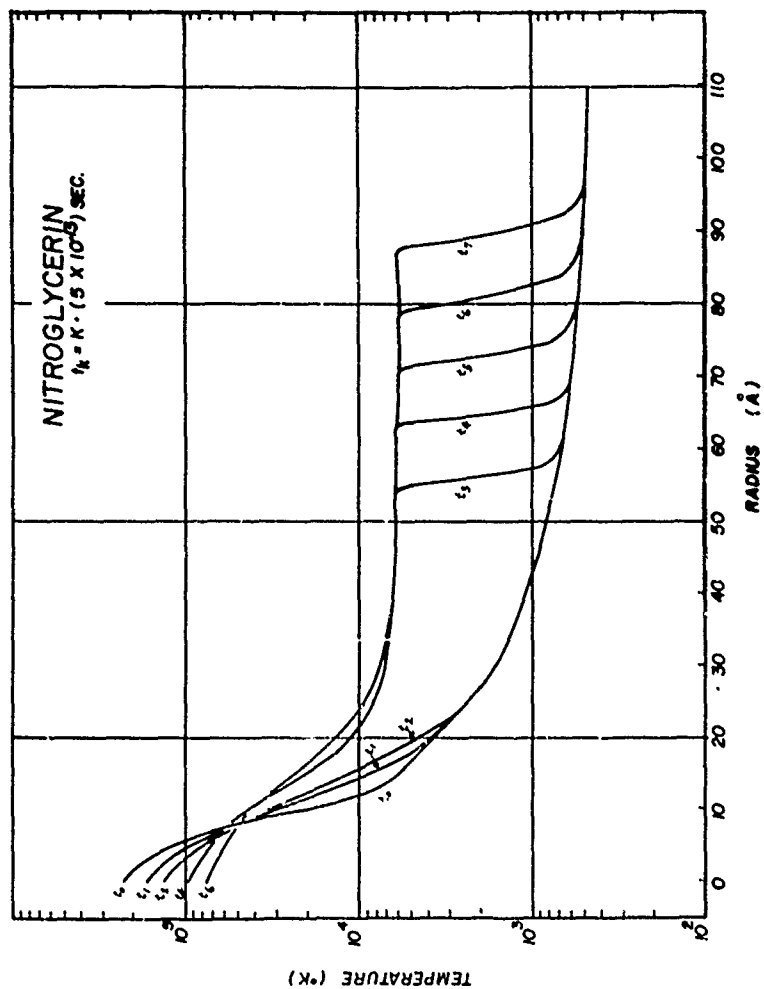


Fig. 2 Temperature vs radius profiles for various times following the passage of the fission-fragment through nitroglycerin. t_k represents a time period of k times the interval 5×10^{-9} seconds

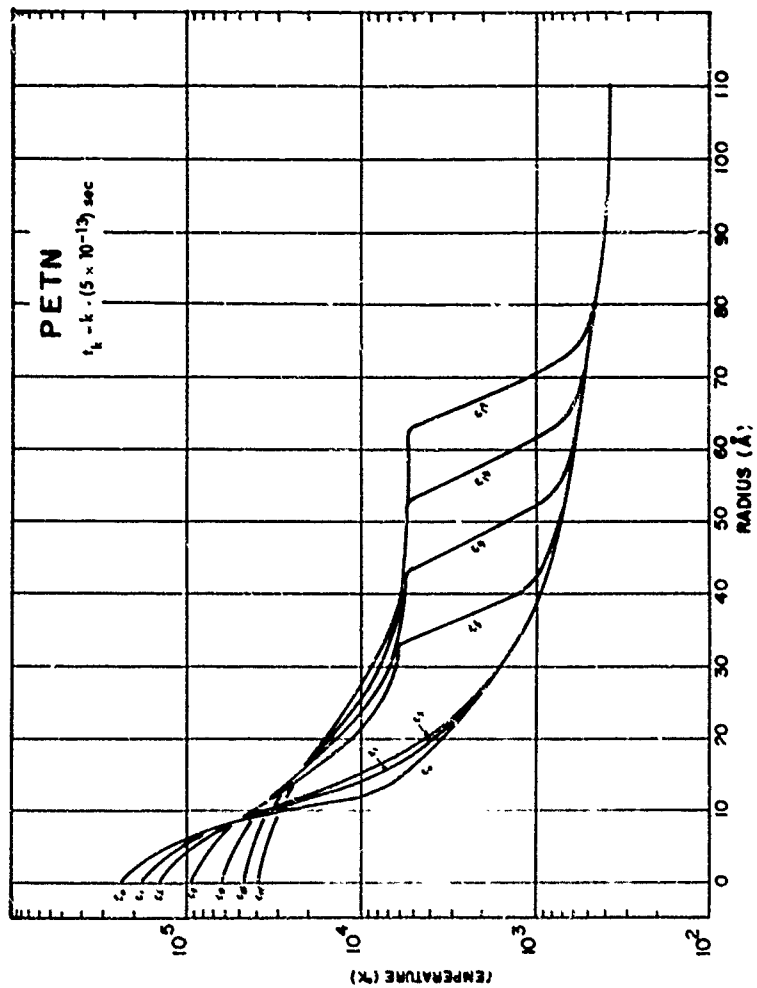


Fig 3 Temperature vs radius profiles for various times following the passage of the fission-fragment through PETN. t_k represents a time period of k times the interval 2×10^{-12} seconds

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INADEQUACY OF THERMAL INITIATION THEORY IN INTERPRETING RESULTS OF FISSION FRAGMENT IRRADIATION OF EXPLOSIVES AT ELEVATED TEMPERATURES

James F. Mallay, Henry J. Prask, Joseph Cerny

Technical Report 3157, June 1964, 19 pp, tables, figures, DA Proj 1C010501A007, ANCMIS Code 5011.11.818A. Unclassified Report

An extreme test of the "hot-spot" model of explosive initiation as applied to microscale irradiation events should occur when the explosives are irradiated at elevated temperatures. To so test this model, fission fragment irradiations of four explosives selected for their high reaction rates (RDX, HMX, PETN, and nitroglycerin) were performed at 125 to 215°C. No

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- 2. Explosives - Initiation
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UNITERMS

Pentaerythritol tetranitrate
Hexahydro-1,3,5-trinitro-s-triazine
Octahydro-1,3,5,7-tetranitro-s-tetrazine
Glyceryl trinitrate
Fission
Fragment
Initiation
Thermal

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